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Excitation functions of deuteron induced nuclear reactions on enriched ^{78}Kr with particular relevance to the production of ^{76}Br

By B. Scholten[1,*], S. Takács[2], F. Tárkányi[2], H. H. Coenen[1] and S. M. Qaim[1]

[1] Institut für Nuklearchemie, Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany
[2] Institute of Nuclear Research of the Hungarian Academy of Sciences, H-4001, Debrecen, Hungary

Dedicated to the memory of Prof. Dr. Dr. h.c. Gerhard L. Stöcklin

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^{78}Kr *gas target* / ^{76}Br / *Excitation function / Integral yield*

Summary. Excitation functions of the ^{78}Kr$(d,\alpha)^{76}$Br, ^{78}Kr$(d,n)^{79}$Rb and ^{78}Kr$(d,x)^{79}$Kr reactions were measured for the first time in the energy range up to 13 MeV using the stacked gas cell technique and activation method. Highly enriched ^{78}Kr was used as target material. The maximum cross section of the ^{78}Kr$(d,\alpha)^{76}$Br reaction at 13 MeV amounts to 21 mb and the calculated yield of ^{76}Br over the energy range $E_d = 13 \rightarrow 4$ MeV to only 59 kBq/μA h. Due to the very low expected yield of ^{76}Br and the high cost of the target gas, this route is not suitable for production of ^{76}Br compared with the other well investigated production routes.

1. Introduction

The radioactive isotopes of bromine with suitable decay characteristics for *in vivo* tracer application are of considerable interest in labelling organic compounds according to the analogue approach. Previously the interest was in ^{75}Br and ^{77}Br; in recent years ^{76}Br is attracting more attention. It is a positron emitter, having a half-life of 16.2 h. The end-point energy of the positrons is fairly high (3.9 MeV), which may be somewhat disadvantageous in investigations using positron emission tomography (PET).

Several reviews have been published on the production of bromine isotopes [cf. 1,2] as well as on the preparation of radiopharmaceuticals labelled with bromine isotopes [cf. 3]. Out of the investigated and potentially useful routes, for the production of ^{76}Br at a small-sized cyclotron, only the ^{76}Se$(p,n)^{76}$Br and ^{78}Kr$(d,\alpha)^{76}$Br reactions are of interest. The cross section data for the ^{76}Se$(p,n)^{76}$Br reaction have been measured [4, 5] and the production method has been technically developed [cf. 6]. Regarding the later reaction using enriched ^{78}Kr, no report was found in the literature. The only work reported on deuteron induced reaction on this target dealt with the yield measurement of ^{75}Br *via* the ^{78}Kr$(d,\alpha n)^{75}$Br reaction from 10 to 21.5 MeV [7].

*Author for correspondence (E-mail: b.scholten@fz-juelich.de).

In the present work we studied several deuteron induced reactions on highly enriched ^{78}Kr in detail. Some preliminary data on the formation of ^{76}Br have already been reported [8].

2. Experimental

The experimental techniques were essentially the same as described in several earlier publications on measurement of cross sections on highly enriched isotopes of krypton [cf. 9–11]. Some details pertinent to the present work are given below.

2.1 Enriched ^{78}Kr target

Two batches of enriched ^{78}Kr were used. Their compositions and purities are given in Table 1. Cylindrical, stainless steel gas cells (20 mm ∅ × 25 mm), filled with enriched ^{78}Kr to a pressure of 0.42–0.92 bar were stacked together (3–5 cells). Polyimide foil (15 μm thick, Kapton) windows were used in irradiations at Debrecen. At Jülich aluminum foils (25 μm and 50 μm thick) were used as windows. Thin titanium foils (12.5 μm and 20 μm) and in some cases copper foils (10 μm) were inserted in front of the stacks and were used as monitors of beam intensity.

2.2 Irradiations and beam current measurement

Irradiations were carried out using the external beam of the compact cyclotron CV 28 in Jülich and the MGC 20E cyclotron in Debrecen. Depending on the primary beam energy several gas cells were stacked and irradiated in a tandem arrangement. In all irradiations only low beam currents (100–200 nA) were used. The beam intensity was monitored by direct collection of the incoming charge in a Faraday cup as well as by using nuclear reactions induced in Ti and Cu foils, which were inserted in front of the gas cells (see above). The cross section data for the monitor reactions ^{nat}Ti$(d,x)^{48}$V and ^{nat}Cu$(d,x)^{65}$Zn used in this work were taken from the literature [cf. 12]. The intensity of the bombarding beam was kept constant during the irradiation

Table 1. Isotopic composition and chemical purity of enriched krypton at delivery.

	Isotopic composition (%)				Chemical purity (%)	Supplier
	^{78}Kr	^{80}Kr	^{82}Kr	^{83}Kr		
Batch 1[a]	99.4	0.6	–	–	99.0	Isotec, USA
Batch 2[b]	99.955	0.045	< 0.001	< 0.001	99.04	Chemotrade, Germany (Russian product)

a: Mainly used at Debrecen. Due to extensive previous work on this gas batch [cf. 11], possibly diluted with some air;
b: Purchased later and most of the measurements at Jülich were done using this gas.

Table 2. Main parameters of the irradiations.

Accelerator	Primary energy (MeV)	Energy range covered (MeV)	Number of cells	Cell window (μm)	Cell pressure (bar)	Irradiation time (min)	Beam current (nA)	Monitor foil (μm)
MGC 20E	9.6	8.1–6.4	4	Kapton (15)	0.47	60	59	Ti (20)
MGC 20E	7.4	5.5–4.0	3	Kapton (15)	0.43	60	58	Ti (20)
MGC 20E	10.0	8.5–6.9	4	Kapton (15)	0.46	60	59	Ti (20)
MGC 20E	9.6	8.0–6.4	4	Kapton (15)	0.44	60	38	Ti (20)
CV 28	14.0	12.8–3.7	5	Al (25)	0.44	60	94	
CV 28	14.1	12.4–6.2	4	Al (50)	0.92	58	166	Ti (12.5) Cu (10)
CV 28	14.1	12.6–7.5	4	Al (25) Al (50)	0.65	60	158	Ti (12.5) Cu (10)
CV 28	9.8	7.8–2.2	4	Al (25)	0.8	60	261	Ti (12.5) Cu (10)
CV 28	14.3	13.4–9.3	5	Al (25)	0.62	60	188	Ti (12.5)

to avoid additional corrections. A summary of all the irradiation conditions is presented in Table 2. The beam intensities obtained *via* the Faraday cup and monitor reaction measurements differed in some cases by 20%. For the calculation of the cross section we used the results obtained *via* monitor reactions.

2.3 Measurement of radioactivity

The activity of each cell was determined without chemical separation *via* standard high-resolution gamma-ray spectroscopy using HPGe detectors. The sources were counted at large distances (0.5 m or larger) from the detector to avoid corrections due to extended source and to reduce the pile up and coincidence effects. On the basis of an independent calibration process it was found that, at the counting distance used, the middle plane of the cell can be regarded as a point source. The estimated relative uncertainty in the detector efficiency was 5%. The measurement was done repeatedly for about 18 to 36 hours to follow the decay of the radioisotopes concerned and for separation of the parent-daughter activities.

The nuclear data for determination of absolute activities from the count rates and for calculation of the decay factors during the irradiations were taken from the literature [cf. 13] and are collected in Table 3. Considering that the measurements after irradiation were started with some delay, the gamma lines of the short half-lived isomers ^{76m}Br (1.31 s), ^{77m}Br (4.3 min), ^{79}Kr (50 s) did not appear in our spectra. Therefore we could determine only the cumulative cross sections for the ground state isomers.

2.4 Determination of cross section

From the experimentally determined beam intensity and the absolute activity of the product (see above) the cross section for the formation of the product was calculated using

Table 3. Decay characteristics of the radionuclides studied and the threshold-energies of the contributing reactions.

Nuclide	Half-life	Mode of decay (%)	E_γ (keV)	I_γ (%)	Main contributing reaction	Threshold energy (MeV)
^{75}Br	1.62 h	EC (29), β^+ (71)	286.6	92	^{78}Kr($d, \alpha n$)	2.4
^{76}Br	16.2 h	EC (46), β^+ (54)	559.1	74	^{78}Kr(d, α)	0.0
			657.1	15.6		
^{79}Kr	1.46 d	EC (92.9), β^+ (7.1)	261.3	12.7	^{78}Kr(d, p)	0.0
			397.5	9.3		
^{79}Rb	22.9 min	EC (18), β^+ (82)	182.8	19.2	^{78}Kr(d, n)	0.0
			688.3	23.1		

the standard activation formula. Two other important factors involved needed careful consideration.

Number of target nuclei

The number of ^{78}Kr nuclei in the cell was determined from the geometric factors of the target gas, the isotopic composition, the target filling pressure, and the actual temperature. The gas cells were irradiated with low beam intensity to avoid gas density reduction along the beam. No special correction was made for the gas density reduction effect.

Primary bombarding energies and energy degradation

The primary beam energies at the MGC 20E cyclotron were determined on the basis of magnetic deflection. At the CV 28 the primary energies were measured by a modified time of flight method [cf. 14]. The middle energies effective at the monitor foils and the gas cells were determined *via* calculation [cf. 15] by using the proper thickness and isotopic compositions of the elements along the degraded beam. The available monitor reactions for low energy deuterons did not allow their effective use for the determination of the energy. Therefore the energy scale of each excitation function is based on well-measured primary projectile energies and carefully measured target parameters.

Uncertainties in cross section data

The uncertainties in the cross sections were determined for each target separately from the estimated uncertainties in the contributing parameters of the activation formula: absolute activity (up to 15%), beam current (10%), nuclear data (3%), number of target nuclei (5%–10%). The resulting uncertainty was calculated by quadratic summation of the individual uncertainties and was found to be in the range 15%–20%.

The uncertainties in the effective projectile energies were calculated by taking into account the cumulative uncertainties, the inhomogeneities in the degrading target elements and the beam broadening due to straggling. Typical estimated uncertainty in the mean energy was about 0.3 MeV at the front foil of the stack. In long stacks the energy uncertainty could reach 0.7 MeV for the last cell.

3. Results and discussion

3.1 Cross section data

The measured cross sections for the formation of a few radionuclides in deuteron induced reactions on ^{78}Kr are given in Table 4. The main contributing processes are listed in

Table 4. Measured cross sections for the formation of a few radionuclides in deuteron induced reactions on ^{78}Kr.

Deuteron energy [MeV]	Formation cross section [mb]			
	^{75}Br	^{76}Br	^{79}Kr [a]	^{79}Rb
4.78 ± 0.5		0.5 ± 0.1		37 ± 7
5.47 ± 0.3		1.5 ± 0.3		
6.00 ± 0.5		3.7 ± 0.6	276 ± 53	
6.33 ± 0.4		4.2 ± 0.7		118 ± 22
6.41 ± 0.5		4.6 ± 0.9		
6.43 ± 0.5		3.3 ± 0.6		
6.91 ± 0.5		5.3 ± 1.0		
7.00 ± 0.5		6.4 ± 1.0		
7.00 ± 0.4		5.7 ± 0.9		
7.46 ± 0.5		6.5 ± 1.2		
7.48 ± 0.5		8.5 ± 1.7	387 ± 71	
7.55 ± 0.4		7.8 ± 1.3		
7.55 ± 0.4		8.1 ± 1.5		
7.77 ± 0.3		9.4 ± 1.6		158 ± 31
7.98 ± 0.4		9.7 ± 1.9		
8.06 ± 0.3		10.5 ± 2.1		
8.08 ± 0.3		9.9 ± 1.9		
8.45 ± 0.5		11.0 ± 1.9	370 ± 74	141 ± 27
8.49 ± 0.3		11.9 ± 2.2		
9.03 ± 0.7		13.3 ± 2.6	506 ± 99	
9.03 ± 0.7		12.2 ± 2.2	386 ± 77	161 ± 32
9.55 ± 0.5		15.4 ± 3.0	384 ± 69	135 ± 27
10.03 ± 0.6		16.8 ± 3.1	512 ± 101	138 ± 26
10.12 ± 0.6		16.5 ± 3.1	337 ± 67	
10.46 ± 0.4		17.9 ± 3.5	325 ± 64	126 ± 22
10.97 ± 0.5		20.2 ± 3.8	412 ± 77	119 ± 23
11.13 ± 0.4	0.56 ± 0.14	20.8 ± 4.2	312 ± 61	
11.32 ± 0.4	0.44 ± 0.11	19.2 ± 3.5	213 ± 40	101 ± 19
11.84 ± 0.4		21.1 ± 4.1	377 ± 69	101 ± 20
12.08 ± 0.4	1.6 ± 0.4	21.1 ± 3.9	283 ± 55	
12.22 ± 0.3	1.8 ± 0.4	20.6 ± 3.8	265 ± 53	89 ± 17
12.59 ± 0.3	2.8 ± 0.6	20.5 ± 3.6	266 ± 47	84 ± 17
12.64 ± 0.3		20.4 ± 3.5	333 ± 58	86 ± 16
13.20 ± 0.3	4.7 ± 1.2	21.6 ± 4.2	235 ± 44	

a: ^{79}Kr is formed independently *via* the ^{78}Kr(p, d) reaction as well as *via* the decay of ^{79}Rb. The cross section given here is a sum of the two processes.

Table 3. The radionuclide ^{79}Kr is formed directly *via* the ^{78}Kr(d, p) reaction as well as *via* the decay of ^{79}Rb. The cross section given in Table 4 is thus a sum of the two processes.

3.2 Excitation functions

The formation of ^{75}Br *via* the ^{78}Kr($d, \alpha n$) reaction was observed only above $E_d = 10$ MeV. The cross section increases with the increasing deuteron energy, reaching a value of 5 mb at 13 MeV. The other investigated processes are discussed below individually.

^{78}Kr(d, α)^{76}Br

The measured cross sections are sums of metastable and ground state cross sections. The data are plotted as a function of deuteron energy in Fig. 1. The reaction starts at about 4.5 MeV and reaches a maximum value of 21 mb at about 13 MeV. Its shape is similar to those of other recently investigated (d, α) reactions in the medium mass region [cf. 16,17]. In general, the cross section of the process is rather low.

^{78}Kr(d, n)^{79}Rb

The measured data are shown as a function of deuteron energy in Fig. 2. The threshold of the reaction lies at about 4 MeV. The cross section increases with energy, reaching a maximum value of about 150 mb at 8.3 MeV. The shape of the excitation function suggests the dominant role of the compound nucleus process. It is similar to those of other recently investigated (d, n) reactions in the medium mass region [cf. 16,18]. This reaction channel is fairly strong.

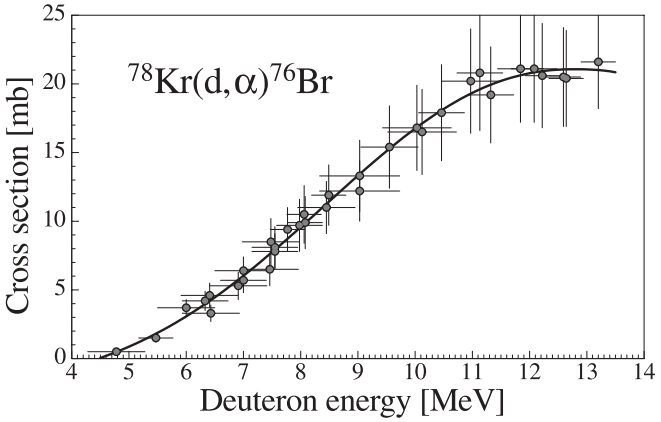

Fig. 1. Excitation function of the ^{78}Kr(d, α)^{76m,g}Br reaction. The solid line is an eye-guide.

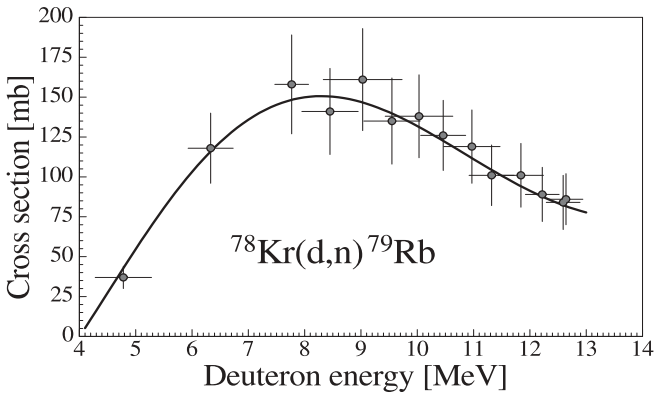

Fig. 2. Excitation function of the ^{78}Kr(d, n)^{79}Rb reaction. The solid line is an eye-guide.

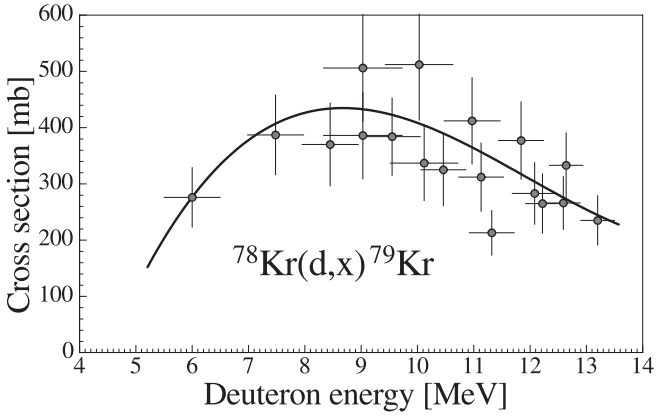

Fig. 3. Cumulative cross section for the formation of ^{79}Kr *via* the ^{78}Kr(d, p)^{79}Kr and ^{78}Kr(d, n)^{79}Rb $\rightarrow$ ^{79}Kr processes plotted as a function of deuteron energy. The solid line is an eye-guide.

^{78}Kr(d, x)^{79}Kr

Regarding the formation of ^{79}Kr, only a sum of the ^{78}Kr(d, p)^{79m,g}Kr and ^{78}Kr(d, n)^{79}Rb $\rightarrow$ ^{79}Kr processes could be determined. The data are plotted as a function of deuteron energy in Fig. 3. The threshold of the process lies at about 4 MeV. The cross section increases with energy, reaching a maximum value of about 450 mb at 9.0 MeV. Comparing Figs. 2 and 3 one comes to the conclusion that the (d, p) channel is much stronger than the (d, n) reaction, an expected corollary in this mass region.

It should be mentioned that a (d, p) reaction leads to the same product as an (n, γ) reaction. An estimate of the secondary neutron flux in our irradiations, and considering that the ^{78}Kr(n, γ)^{79}Kr reaction has a thermal cross section of 6 barns, we estimated the contribution of the (n, γ) process to the formation of ^{79}Kr as negligibly small ($< 1\%$) in comparison to that of the (d, p) reaction.

3.3 Calculated yield of ^{76}Br

From the excitation function of the ^{78}Kr(d, α)^{76}Br reaction given in Fig. 1, the differential and integral yields of ^{76}Br were calculated for an 1 h irradiation at 1 μA. The results on the integral yield are given in Fig. 4. Evidently the expected yield of ^{76}Br is low; for the whole energy range $E_d = 13 \rightarrow 4$ it amounts only to 59 kBq (1.6 μCi)/μA h.

3.4 Evaluation of production possibility

In connection with the production of 75 Br at a small-sized cyclotron the ^{78}Kr(p, α)^{75}Br reaction, originally suggested by Friedman *et al.* [19], was recently investigated in detail. Extensive cross section measurements [11] and development of a high-current spherical target [20] have shown that ^{75}Br can be produced in quantities sufficient for local use. Using the same target gas and a deuteron beam, with the aim to produce ^{76}Br for local use *via* the ^{78}Kr(d, α) reaction, how-

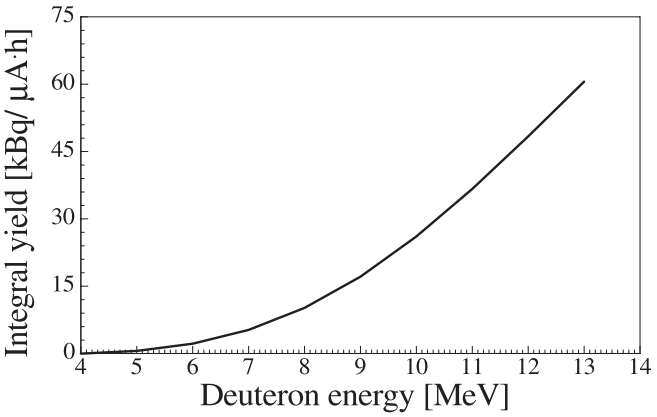

Fig. 4. Integral yield of ^{76}Br calculated from the excitation curve given in Fig. 1, plotted as a function of deuteron energy.

ever, does not fulfil the expectation. The yield of ^{76}Br is so low that it is not meaningful to pursue this route further; particularly in view of the fact that the highly enriched ^{78}Kr is rather expensive, and the other methods of ^{76}Br production already in use are more efficient. The same is true for the other two investigated radionuclides, *viz.* ^{79}Rb and ^{79}Kr. If needed they can be produced *via* more efficient methods than the *d*-induced reactions on ^{78}Kr described here.

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